Generation and Application of Femtosecond X-Rays from a Synchrotron

R.W. Schoenlein^a, H.H.W. Chong^b, T.E. Glover^c P.A. Heimann^c, C.V. Shank^a, A.A. Zholents^d, and M.S. Zolotorev^d

^a Materials Sciences Division, Ernest Orlando Lawrence Berkeley National Laboratory
 1 Cyclotron Rd. MS: 2-300, Berkeley CA 94720, email: rwschoenlein@lbl.gov
^b Applied Science and Technology Graduate Group, University of California Berkeley, Berkeley CA 94720
 ^c Advanced Light Source Division, Lawrence Berkeley National Laboratory
^d Accelerator and Fusion Research Division, Lawrence Berkeley National Laboratory

An important new area of scientific research in chemistry, physics, and biology is the investigation of ultrafast structural dynamics in condensed matter using femtosecond x-ray pulses. X-rays are powerful probes of atomic structure since they interact with core electronic levels, and can therefore provide direct information about relative atomic positions and coordination. Modern synchrotrons have proven to be powerful tools for probing the "static" structure of matter. Techniques such as x-ray diffraction, extended x-ray absorption fine structure (EXAFS) and many others are widely used at such facilities to obtain structural information with atomic resolution. However, the time resolution provided by synchrotrons is limited to >30 ps due to the length of the stored electron bunches. This is orders of magnitude longer than the fundamental time scale for atomic motion, dictated by an atomic vibrational period, ~100 fs.

We demonstrate a novel scheme for generating ultrashort pulses of synchrotron radiation[1]. Our approach is to create femtosecond time-structure on a long electron bunch by using a femtosecond laser pulse to modulate the energy of an ultrashort slice of the bunch. Femtosecond synchrotron pulses are directly measured from a bend-magnet beamline at the Advanced Light Source (ALS)[2, 3].

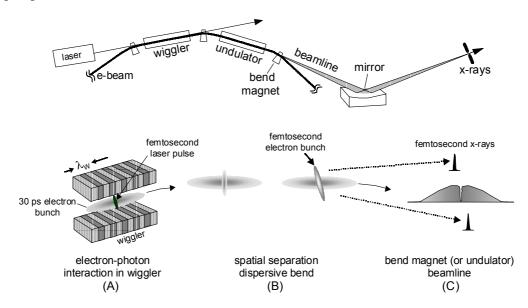


Fig. 1. Schematic of method for generating femtosecond synchrotron pulses. (A) Femtosecond laser pulse interaction with the electron bunch in a permanent-magnet wiggler. (B) Transverse separation of modulated electrons in a dispersive bend of the storage ring. (C) Generation of synchrotron radiation from a bend-magnet (or undulator) and separation of the femtosecond synchrotron radiation at the beamline image plane.

Figure 1 illustrates the modulation and generation scheme. A femtosecond laser pulse copropagates with the stored electron bunch through a wiggler (Fig. 1A). The high electric field of the laser pulse modulates the energy of the underlying electrons as they traverse the wiggler. The optimal interaction occurs when the central wavelength of the spontaneous emission from an electron passing

through the wiggler matches the laser wavelength (FEL resonance condition)[1]. In addition, the transverse mode of the laser beam must match the transverse mode of the spontaneous radiation from the electron passing through the wiggler, and the laser spectral bandwidth must match the spectrum of the fundamental wiggler radiation averaged over the transverse mode.

By creating an energy modulation that is significantly larger than the beam energy spread, the transverse dispersion of the storage ring will cause a spatial displacement of the modulated electrons from the rest of the electron bunch (Fig. 1B). Finally, by imaging the synchrotron radiation from the displaced electrons to the experimental area, femtosecond x-rays can be separated from the long-pulse using an aperture (Fig. 1C). The time structure of the temporally incoherent synchrotron radiation is directly determined by the time structure of the electron bunch and is invariant over the entire spectrum of the synchrotron emission, from infrared to x-ray wavelengths. We directly measure the time-structure of the visible synchrotron pulses via cross-correlation with a delayed pulse from the laser system. An adjustable knife-edge located in the beamline at an intermediate image plane provides a means to select synchrotron radiation originating from different transverse regions of the electron beam.

Figure 2 shows two cross-correlation measurements made at various knife-edge positions, in units of the rms horizontal beam size, σ_x . Measurements of the central core of the synchrotron beam (Fig. 2A) reveal the femtosecond hole or dark pulse that is created due to the acceleration (and deceleration) of electrons by the laser pulse, and their subsequent transverse spatial separation from the central core of the bunch. The bright pulse is measured with the knife-edge at the $3\sigma_x$ position (Fig. 2B) such that the central core of the beam is blocked. The solid lines in Fig. 2 are predicted pulse shapes based on a model calculation of the electron bunch distribution (using known parameters of the electron beam and the storage-ring lattice) [3].

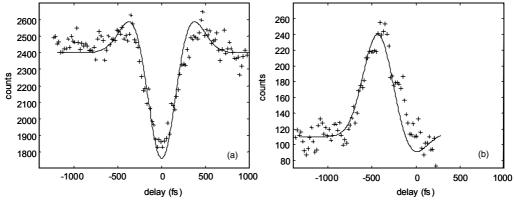


Fig. 2. Cross-correlation measurements between a delayed laser pulse and synchrotron radiation originating from an energy-modulated electron bunch. In (A), synchrotron radiation from the central core $(\pm 3\sigma_x)$ of the electron bunch is selected. In (B), synchrotron radiation from the horizontal wings $(\pm 3\sigma_x)$ to $\pm 8\sigma_x$ of the electron bunch is selected. Solid lines are from a model calculation[3].

The measured pulse duration is determined by the storage ring dispersion integrated from the wiggler to the radiation bend-magnet. A new beamline, dedicated to ultrafast x-ray spectroscopy is currently under construction at the ALS and will provide x-ray pulses of <100 fs duration. The femtosecond time structure is invariant over the entire spectral range of bend-magnet emission from the near infrared to the x-ray regime, making this a powerful tool for femtosecond x-ray spectroscopy.

Supported by the Director, Office of Science, Office of Basic Energy Sciences, of the U.S. Department of Energy under Contract No. DE-AC03-76SF00098.

References

- 1. A.A. Zholents, M.S. Zolotorev, *Phys. Rev. Lett.* **76**, 912, 1996.
- 2. R.W. Schoenlein, S. Chattopadhyay, H.H.W. Chong, T.E. Glover, P.A. Heimann, C.V. Shank, A. Zholents, and M. Zolotorev, *Science*, **287**, 2237, 2000.
- 3. R.W. Schoenlein, S. Chattopadhyay, H.H.W. Chong, T.E. Glover, P.A. Heimann, C.V. Shank, A. Zholents, and M. Zolotorev, *Appl. Phys. B*, **71**, 1, 2000.